

Large upper critical field in non-centrosymmetric superconductor Y_2C_3

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Abstract

We determine the upper critical field $\mu_0 H_{c2}(T_c)$ of non-centrosymmetric superconductor Y_2C_3 using two distinct methods: the bulk magnetization $M(T)$ and the tunnel-diode oscillator (TDO) based impedance measurements. It is found that the upper critical field reaches a value of 30T at zero temperature which is above the weak-coupling Pauli paramagnetic limit. We argue that the observation of such a large $\mu_0 H_{c2}(0)$ in Y_2C_3 could be attributed to the admixture of spin-singlet and spin-triplet pairing states as a result of broken inversion symmetry.

Keywords: Superconductor, critical phenomena, Thermodynamic properties,

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1. Introduction

Among the previously investigated superconductors, most of them possess a center of inversion in their crystal structure. In these cases, the Pauli principle and the parity conservation then dictate that the superconducting pairing states with even parity are necessarily spin singlet, while those with odd parity must be spin triplet [1, 2]. However, such a tie between the spatial symmetry and the Cooper-pair spin may be broken in the non-centrosymmetric superconductors in which the inversion symmetry is absent [3, 4, 5, 6].

The recent discovery of superconductivity in CePt_3Si [7] has largely stimulated the investigation of superconductivity lacking inversion symmetry. In the past few years, a growing number of non-centrosymmetric superconductors have been successfully synthesized. Evidence for the admixture of spin-singlet and spin-triplet components has been found in $\text{Li}_2(\text{Pd,Pt})_3\text{B}$ [8, 9, 10, 11] and the heavy fermion systems of CePt_3Si [12, 13, 14], CeRhSi_3 [15, 16] and CeIrSi_3 [17, 18, 19]. In $\text{Li}_2(\text{Pd,Pt})_3\text{B}$, it was shown that the spin triplet state might become dominant with enhancing the antisymmetric spin-orbit coupling (ASOC) strength [8, 9]. On the other hand, the superconducting pairing state might become complicated by strong electron correlations in the heavy fermion compounds. Therefore, materials having weak electronic correlations are desired for studying the effect of ASOC on superconductivity.

Y_2C_3 is a non-centrosymmetric superconductor with a superconducting transition temperature T_c of up to 18K, the highest among all the investigated non-centrosymmetric superconductors [20]. It crystallizes in a body-centered cubic structure (Pu_2C_3 -type) with space group $\bar{I}43d$. Until now, only a very

few physical properties have been studied for Y_2C_3 . Measurements of specific heat indicates an isotropic superconducting gap of s-wave type [21], but the NMR [22] and μSR [23] experiments support a scenario of multi-gap superconductivity. The upper critical field $\mu_0 H_{c2}(0)$ derived from the initial slope at T_c is estimated to be over 30T [24], which is close to the Pauli paramagnetic limit. Such a large upper critical field might indicate the significant contribution of spin triplet component to the pairing state. To determine the true value of the upper critical field and its temperature dependence, one needs extend measurements to much higher magnetic fields.

In this article, we report the upper critical field $\mu_0 H_{c2}(T_c)$ of Y_2C_3 measured by magnetization $M(T)$ up to 14T and by the TDO resonant circuit using the facilities of pulsed magnetic field at Los Alamos. An upper critical field of about 30T is obtained with T_c of 15.5K. We argue that the contributions of spin triplet state resulting from the parity violated ASOC might lead to the enhancement of the upper critical field.

2. Experimental methods

Polycrystalline samples were prepared by mixing appropriate amounts of Y (99.9%) and C (99.95%) powders in a dry box [21]. The binary alloys were first prepared by arc melting, after which they were subjected to high-pressure and high-temperature treatment to produce the sesquicarbide phase. All subsequent sample treatments, including preparations for high-pressure experiments, were performed in a dry helium atmosphere. The melted samples were heated to 1400 - 1600 °C in 5 minutes and then maintained at this temperature for 30 minutes under a pressure of 4.0 - 5.0 GPa using a cubic-anvil high-pressure apparatus. After that, the samples were quickly quenched to room temperature within a few seconds. The achieved ingots were examined by powder x-ray diffrac-

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tion using a conventional X-ray spectrometer with a graphite monochromator, which confirmed Y_2C_3 as a single phase.

Since the samples of Y_2C_3 are air sensitive, one needs avoid exposing the samples in air. In order to minimize the time for setting up the experiments, we choose the contactless measurements of magnetization and the TDO resonant frequency (see below) to determine the upper critical field. The magnetization was measured in a commercial Quantum Design magnetic properties measurement system (MPMS) with a magnet field up to 14T. In order to measure $\mu_0 H_{c2}$ down to zero temperature, we performed experiments in pulsed magnetic fields up to 35T at Los Alamos, using a TDO-based technique in which two small counter-wound coils form the inductance of a resonant circuit. The resonant frequency (about 31M Hz) can depend on both the skin-depth (or, in the superconducting state, the penetration depth) and the differential magnetic susceptibility of the sample. A piece of thin sample (about $0.8 \times 0.6 \times 0.1 \text{ mm}^3$) was inserted in the coils, which were immersed in ^3He liquid or ^3He exchange gas, temperatures being measured with a Cernox thermometer 5 mm away from the sample.

3. Results and Discussion

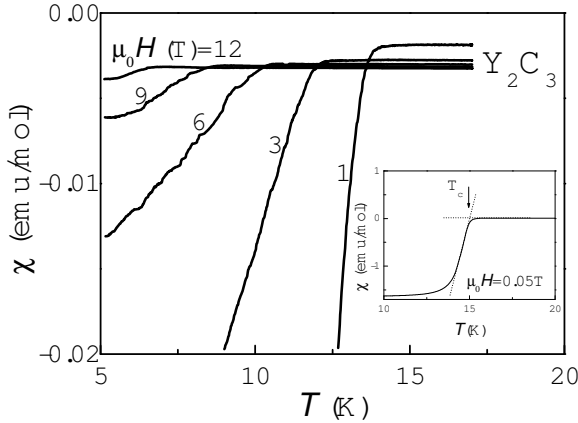


Figure 1: Temperature dependence of the magnetic susceptibility $\chi(T)$ at variant magnetic fields. T_c is determined from the onset of the superconducting transition as shown in the inset.

Fig.1 presents the temperature dependence of the magnetic susceptibility $\chi(T)$ of Y_2C_3 at selected magnetic field. To clearly show the superconducting transition at all the magnetic fields, we only plot the superconducting transition partially for the small fields. A rather sharp superconducting transition with $T_c \approx 15\text{K}$ is observed at low field (see the inset), indicating good sample quality. With increasing magnetic field, the superconducting transition is shifted to lower temperature, but superconductivity is not yet suppressed at a field as high as 14T. Therefore, facilities with higher accessible magnetic field, e.g., the pulsed magnet, is desired in order to track the critical field down to zero temperature. From Fig. 1, one can find that the superconducting transition becomes broader and the diamagnetic signal is weakened upon applying a magnetic field which

is likely attributed to the flux pinning effect. In the context here, the superconducting transition temperatures T_c^χ are determined from the onset of the superconducting transition as shown in the inset of Fig. 1.

In order to further suppress superconductivity down to zero temperature, we have measured the TDO resonant frequency of Y_2C_3 up to 35T using a short pulse magnet. Fig. 2 shows the relative TDO frequency, $\Delta f(\mu_0 H)$, as a function of magnetic field at selected temperatures for Y_2C_3 . The sudden increase of $\Delta f(\mu_0 H)$ upon cooling down marks the onset of the superconducting transition. At temperatures above T_c ($\approx 15\text{K}$), the TDO frequency increases smoothly with decreasing temperature. The critical field of $\mu_0 H_{c2}^{TDO}$ is determined from the onset of the transition as described in the inset of Fig. 2. A magnetic field of about 30T is required to completely suppress superconductivity in Y_2C_3 as seen in Fig.2. The successful measurement of superconductivity and its upper critical field $\mu_0 H_{c2}(T_c)$ in Y_2C_3 indicates that the TDO-based impedance measurement is a very powerful technique in studying air-sensitive materials.

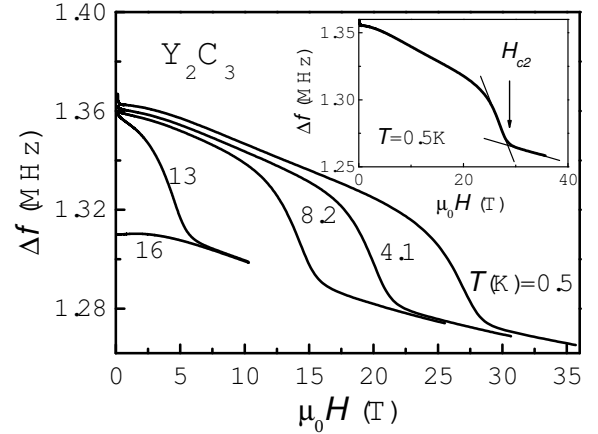


Figure 2: Magnetic field dependence of the resonant frequency shift $\Delta f(H)$ at temperatures of $T = 0.5\text{K}$, 4.1K , 8.2K , 13K and 16K . The inset shows $\Delta f(H)$ at $T = 0.5\text{K}$, in which the critical field $\mu_0 H_{c2}$ is determined from the intersection point.

The upper critical fields $\mu_0 H_{c2}(T_c)$, obtained from the magnetic susceptibility $\chi(T)$ and the TDO resonant frequency $\Delta f(H)$ as described above, are plotted as a function of temperature in Fig.3. The discrepancy between $\mu_0 H_{c2}^\chi(T_c)$ and $\mu_0 H_{c2}^{TDO}(T_c)$ might originate from the fact that the magnetic susceptibility measures the bulk superconductivity while the TDO resonant technique is a kind of surface measurement. Nevertheless, the two curves of $\mu_0 H_{c2}(T_c)$ follow qualitatively similar behavior: $\mu_0 H_{c2}$ shows a weak upturn close to T_c , and then increases linearly with decreasing temperature. The extension of $\mu_0 H_{c2}(T_c)$ to zero gives $T_c^\chi = 15\text{K}$ and $T_c^{TDO} = 15.5\text{K}$, respectively. Two important features can be observed in Fig. 3: (i) Y_2C_3 possesses a large value of upper critical field ($\mu_0 H_{c2}^\chi(0) \approx 30\text{T}$) and (ii) $\mu_0 H_{c2}(T_c)$ shows very unusual temperature dependence.

In a conventional BCS superconductor, two electrons with opposite spins and momenta form a Cooper pair. Application

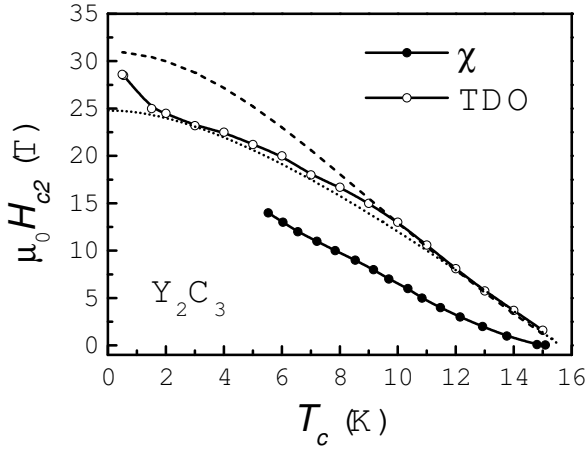


Figure 3: The upper critical field versus temperature for Y_2C_3 . The filled and open symbols represent the data derived from the magnetization and the relative TDO resonant frequency, respectively. The dotted line and the dashed line are fits to the weak coupling WHH theory and $\mu_0 H_{c2}(T_c) = \mu_0 H_{c2}(0) \frac{1-(T/T_c)^2}{1+(T/T_c)^2}$, respectively.

of a magnetic field can break the Cooper pairs and, therefore, suppress superconductivity via the following two channels: (1) the orbit channel in which the charges with opposite momenta are decoupled via Lorentz force in a magnetic field (i.e., the so-called orbital limit), and (2) the spin channel is broken via the Zeeman effect in which the spin singlet is transferred to spin triplet. At sufficiently high magnetic field, superconductivity can be destroyed by orbital and spin pair breaking. However, for a triplet superconductor, the spin channel is robust against magnetic field and, therefore, its upper critical field can be largely enhanced in comparison with singlet superconductors [25].

According to the Werthamer-Helfand-Hohenberg (WHH) method [26], the orbital limited upper critical field is given by:

$$\mu_0 H_{c2}^{orb}(0) = -0.69 T_c (d\mu_0 H_{c2}/dT)_{T_c}. \quad (1)$$

For a weak-coupling BCS superconductor, the Pauli paramagnetic limiting field can be expressed as [27, 28]:

$$\mu_0 H_{c2}^P(0) [\text{Tesla}] = 1.86 T_c [\text{K}]. \quad (2)$$

For Y_2C_3 , the superconducting transition temperature and the initial slope of the upper critical field are $T_c^{TDO} = 15.5$ K and $(d\mu_0 H_{c2}/dT)_{T_c} = -2.3$ T/K, respectively. From Eq. (1) and Eq. (2), one can then calculate the upper critical fields associated with the orbital limit and the spin Pauli paramagnetism, which gives $\mu_0 H_{c2}^{orb}(0) = 24.5$ T and $\mu_0 H_{c2}^P(0) = 28.8$ T. Therefore, our experimentally derived value of $\mu_0 H_{c2}^{TDO}(0) = 30$ T is larger than those obtained within the BCS theory. Furthermore, $\mu_0 H_{c2}(T_c)$ shows upturn curvatures close to T_c and also at low temperature, deviating from the predictions of the WHH theory [26] and the Ginzburg-Landau theory [29] in which $\mu_0 H_{c2}(T_c)$ usually gets flat as the temperature goes to zero. Both the large upper critical field $\mu_0 H_{c2}(0)$ and the upturn curvature in

$\mu_0 H_{c2}(T_c)$ are typically found in non-centrosymmetric superconductors with a large antisymmetric spin-orbital coupling [16, 18]. However, we still couldn't rule out the possibilities that the upper critical field might be enhanced by strong coupling. To find out whether these unique properties, including the large upper critical field and its unusual temperature dependence, are attributed to the spin-triplet state as a result of ASOC effect in non-centrosymmetric superconductors, further measurements are desired in order to elucidate the order parameter symmetry. The recent NMR [22] and μ SR [23] measurements claimed that Y_2C_3 is a two-band superconductor like in MgB_2 [30]. However, these measurements have been only performed down to 2K and its data resolution is poor at low temperature. Thus it is difficult to fit their low temperature behavior precisely. Further lower temperature measurements, e.g., the penetration depth, specific heat and NMR, are, therefore, highly desired. One possible scenario may happen as follows: the contributions of spin-triplet state and spin-singlet states are comparable in Y_2C_3 , nodes may only develop at very low temperature and H_{c2} can be enhanced, as we see here, due to the contribution of spin-triplet state.

4. Conclusion

In summary, we have determined the upper critical field $H_{c2}(T_c)$ of Y_2C_3 by measuring the magnetization and the TDO-based resonant frequency. We found that Y_2C_3 possesses a large upper critical field of $\mu_0 H_{c2}(0) \approx 30$ T, exceeding the spin paramagnetic limit in weak coupling. Furthermore, an upturn curvature is observed in $\mu_0 H_{c2}(T_c)$ at low temperatures. These unusual magnetic properties might highlight the importance of broken inversion symmetry on the superconducting properties of Y_2C_3 , in which the contributions from spin-triplet state might play a role.

5. Acknowledgements

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